Measurement of hydrogen depth distribution by resonant nuclear reactions[a)]

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The resonance at $E(^{19}F) = 6.42$ MeV in the reaction $^{1}H(^{19}F,\alpha\gamma)^{16}O$ has been explored as a potentially useful method for the quantitative determination of hydrogen concentration as a function of depth in a solid substrate. The relative merits of this resonance, the 16.44-MeV resonance in the same reaction, and the 6.39-MeV resonance in the reaction $^{1}H(^{15}N,\alpha\gamma)^{12}C$ are discussed.

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The importance of being able to determine quantitatively the depth distribution of hydrogen implanted in various materials, or produced within the materials by fast neutron bombardment, has been emphasized recently by Vook *et al.*[1] in the context of present and future energy producing reactors. It appears that the resonant nuclear reaction technique will be useful in this kind of determination. The use of the $E(^{19}F) = 16.44$-MeV resonance in the reaction $^{1}H(^{19}F, \alpha\gamma)^{16}O$ as a sensitive probe of the depth distribution of hydrogen was reported first by Leich and Tombrello,[2] who employed this technique to profile solar-wind-implanted hydrogen in Apollo lunar soil samples (see also Ref. 3). The same resonance was also employed in measurements of the thickness of the hydration layers of obsidian artifacts, as an aid to dating these artifacts.[4] In Ref. 2 the usefulness of the narrow $E(^{15}N) = 6.385$-MeV resonance in the reaction $^{1}H(^{15}N, \alpha\gamma)^{12}C$ as a method for studying hydrogen depth profiles with better resolution for shallow depths was pointed out, and the use of this resonance has been reported recently by Lanford *et al.*[5] The object of the present letter is to point out the attractive features of the $E(^{19}F) = 6.42$-MeV resonance in the $^{1}H(^{19}F, \alpha\gamma)^{16}O$ reaction as yet another tool for the study of hydrogen depth distributions. We discuss the relative merits of the three resonances for hydrogen depth profiling and the limitations caused by the degradation of resolution that results from energy-loss straggling.

In the present work, ^{19}F beams from the ONR-CIT tandem accelerator were magnetically analyzed and directed to the target. Beam currents of approximately 300 nA (ion charge state +3) were integrated for approximately 100 sec to give an integrated charge of 30 μC per point on the yield curve. The 6- and 7-MeV γ rays deexciting the residual ^{16}O nuclei[6] were detected by an unshielded 12.7-cm-diam 10.2-cm-thick NaI(Tl) scintillator. Since the γ rays are isotropic[7] at the $E(^{19}F) = 6.42$-MeV resonance, the scintillator was place at 0°, with the front face 1.5 cm from the target. At each bombarding energy, a pulse-height spectrum was accumulated in a multichannel analyzer, and the yield was taken as the number of counts in a pulse-height window from 3.5 to 8.0 MeV. With the detector geometry and energy window used, the fraction of all γ rays emitted which are recorded is estimated to be 0.13 (Ref. 8). Other details of procedure are similar to those of published hydrogen depth-profile measurements.[2–5]

Figure 1 shows the γ-ray yield as a function of bombarding energy, using the 6.42-MeV ^{19}F resonance as a probe for the hydrogen contained in a 2800-Å-thick beryllium layer, evaporated onto a spectroscopically pure copper backing at a pressure of 4×10^{-6} Torr. In the experiment for which these measurements were made, the γ-ray yield measurements were used to determine the thickness of the beryllium layer,[9] as well as to check on the hydrogen content of the layer. The hydrogen content was determined in absolute terms by comparison with the yield of γ rays obtained by bombarding layers of the commercial materials Kapton and Lexan, in which the hydrogen content is known. An average hydrogen concentration within the beryllium film of 0.6 at.% was determined. The surface peak

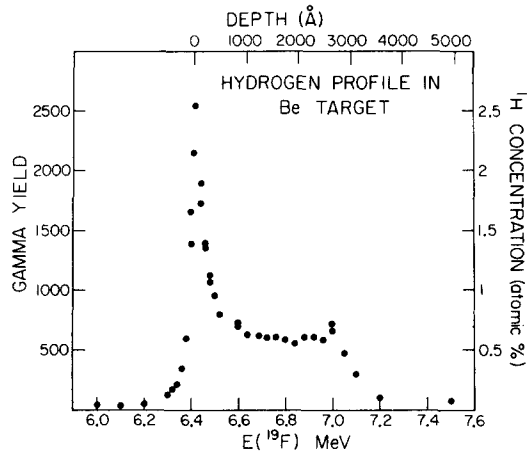

FIG. 1. Profile of hydrogen distribution in a 2800-Å beryllium film deposited on a copper substrate. The ordinate represents the integrated number of events in a γ-ray pulse-height window of $3.5 < E_\gamma < 8.0$ MeV, normalized to a fluence of 6×10^{13} ^{19}F ions. The surface hydrogen peak has a width at half-maximum of 50 keV, in agreement with the width of 45 keV expected from Table I. The background counts below 6.2 and above 7.2 MeV could be reduced substantially by shielding the detector with several inches of lead.

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TABLE I. Parameters of interest in hydrogen depth-profiling.

	$^1H(^{19}F,\alpha\gamma)^{16}O$ [a]		$^1H(^{15}N,\alpha\gamma)^{12}C$ [b]
Resonance energy (MeV)	16.440 ± 0.004	6.418 ± 0.001	6.385 ± 0.015
γ-ray energy (MeV)	6.13, 6.92, 7.12	6.13, 6.92, 7.12	4.44
Resonance width (keV)	88.6 ± 3.8	45.2 ± 3.8	13.4
Relative integrated resonance yield[c]	10.5	1	~1
Depth resolution at the surface of nickel layer (Å)[d]	86	48	18
Depth resolution at 1 μm depth in nickel (Å)[d]	205	190	185
Depth probed in nickel before onset of next resonance (μm)[d]	0.23	0.60	1.80

[a] $^1H + ^{19}F$ resonance parameters were taken from Ref. 6.
[b] $^1H + ^{15}N$ resonance parameters were taken from Ref. 6.
[c] This is proportional to the height of the step in the γ-ray yield measured for a thick hydrogen-containing target, and is therefore proportional to the ultimate sensitivity for measuring hydrogen. For orientation purposes, the $E(^{19}F) = 16.44$-MeV resonance has been used to measure hydrogen contents as low as 20 parts per million in lunar samples. [D. A. Leich, Ph. D. thesis (California Institute of Technology, Pasadena, Calif., 1973) (unpublished); see also Refs. 2 and 5].
[d] Energies were converted to depths with the use of the stopping power and range tables of L. C. Northcliffe and R. F. Schilling [Nucl. Data Tables A 7 (1970)].

evident in Fig. 1 is attributed to the presence of water adsorbed on the target.

Prior to these measurements, an attempt was made to determine the hydrogen content by using the $E(^{19}F) = 16.44$-MeV resonance,[2–4,9] but the yield of γ rays from nuclear reactions between the ^9Be and ^{19}F nuclei was so high that the yield of γ rays from $^1H + ^{19}F$ could not be extracted unambiguously. It seems likely that a similar situation would obtain whenever the layer to be probed for hydrogen also contains significant quantities of elements with atomic number $Z \lesssim 5$. It is probable that the $E(^{15}N) = 6.385$-MeV resonance could also be used successfully in the presence of low-Z materials in the sample to be studied. Deuterium, in amounts comparable to the hydrogen in the target under study, would seriously reduce the sensitivity of the hydrogen measurements for either fluorine or nitrogen beams, because of γ rays from $(d,p\gamma)$ and $(d,n\gamma)$ reactions.

We now turn to a discussion of the relative merits of the three resonances for hydrogen depth profiling. Table I presents a summary of the parameters of interest for determining hydrogen in a layer of nickel. Clearly the $E(^{19}F) = 16.44$-MeV resonance provides the highest sensitivity, because of its large value of $\sigma\Gamma$, the product of resonant cross section and width (line 4 of Table I). Radiation damage of the material during the depth-profile measurements is also least for the higher fluorine resonance, because of this higher yield and lower nuclear stopping power. On the other hand, it has the poorest depth resolution, particularly near the surface of the sample, and the smallest depth of study before the next higher resonance in the reaction begins to produce γ rays at the surface of the sample. The $E(^{15}N) = 6.385$-MeV resonance is superior on both of the latter points of comparison, but it requires that the ion source of the accelerator be operated with gas containing a high percentage of the rare isotope of nitrogen, whereas ^{19}F is the only stable isotope of fluorine. All three resonances can be achieved easily with most tandem accelerators, and fluorine beams are particularly easy to produce in tandem accelerators. A single-ended accelerator with a maximum terminal energy of 6.5 to 7 MV could reach the lower fluorine resonance and the nitrogen resonance with singly-charged ions, and an accelerator with one-half this maximum terminal voltage would probably produce sufficient doubly-charged beam to employ these resonances for depth profiling of hydrogen.

The depth resolution that can be obtained with these resonances depends, of course, on the depth within the sample because of energy-loss straggling. We define the depth resolution as

$$\Delta(x) = \tfrac{1}{2}[\Gamma^2 + (\Delta E)^2]^{1/2}\left(\frac{dE}{dx}\right)^{-1}, \tag{1}$$

where Γ is the full width at half-maximum of the resonance, ΔE is the energy-loss straggling at the depth x, and dE/dx is the (energy-dependent) rate of energy loss in the material under study. Following Bohr,[10] we write

$$\Delta E = 2.355\,(4\pi e^4 z^2 Z N x)^{1/2}, \tag{2}$$

where e is the electronic charge, z and Z are the atomic numbers of the projectile and target, respectively, and N is the number of target atoms per unit volume.

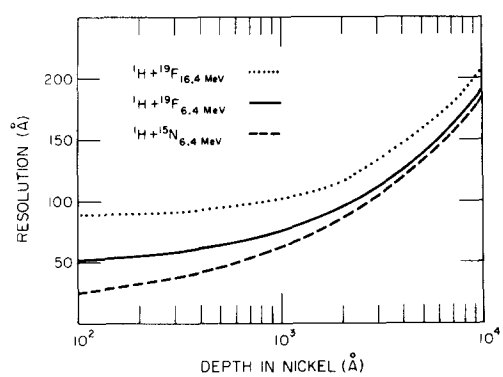

FIG. 2. Semilogarithmic plot of the degradation of resolution by beam straggling. The curves are the results of calculations with Eqs. (1) and (2) for a nickel target. The resolution at the surface is determined only by the resonance width if the spread in the bombarding beam energy is negligible, as has been assumed here.

Although the experimental information on heavy-ion straggling leaves much to be desired, Eq. (2) will be sufficiently accurate to permit a comparison of depth resolution at each of the resonances. Figure 2 shows $\Delta(x)$ as a function of depth in a sample of nickel for the three resonances discussed here. The decided resolution advantage of the ^{15}N reaction at small depths decreases with increasing depth, and is relatively unimportant for depths greater than 0.2 μm.

The best choice among the three resonances for hydrogen depth-profiling studies clearly depends on the chemical constitution of the sample, on the depth to be probed, and on the accelerator which is available for the study. The two low-energy resonances make it possible to carry out depth profiling with relatively low-energy (3.5 MV) accelerators by using doubly charged beams from the ion source.

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ERRATA

Erratum: Energy threshold effects in the collisionless dissociation of polyatomic molecules by ir laser radiation [Appl. Phys. Lett. 30, 514 (1977)]

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A manuscript error was made in line 8, second column, p. 514. $1/e$ should read $\frac{1}{2}$. In addition, we would like to point out an apparent lack of uniformity in the literature in calculating laser intensities and energy densities from measured energies and beam radii. In our work, the $\frac{1}{2}$ beam energy radius was used in area calculations. For our beam, using the the $1/e$ intensity radius gives an area 1.65 times larger and therefore energy densities which are correspondingly smaller.